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INFRARED RADIATION MEASUREMENTS OF COMBUSTION GASES

by

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SUMMARY

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The fabrication of alumina tube window holders and their installation in the high-temperature infrared absorption system is described. Measurements of the temperature distribution along the test optical path with these holders showed the variation to be reduced to 80 K for a center temperature of 1200 K. Absorption measurements of CO at 1200 °K and pressures of 0.25 to 3 atmospheres are presented. These are successfully correlated by the strong line approximation in the Elsasser band model.

Author

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INTRODUCTION

Knowledge of the infrared radiative properties of rocket exhaust gases is required in many applications. Examples include the prediction of radiative heating to the base regions of multi-engine vehicles and the long-range detection of ballistic missiles. The magnitude and characteristics of this infrared radiation depends on the flame composition and temperature.

Analytical studies of the radiative heating from rocket exhaust plumes indicated the need for complete and accurate spectral absorption data. Although numerous investigations of CO_2 and H_2O have been reported recently, the results are for a maximum temperature of 2000°F or are restricted to a narrow wavelength region. Further more, existing theories for predicting infrared radiation from high temperature gases are based on highly idealized physical models. The validity and applicability of the results predicted from these theories require experimental conformation.

The objective of the present study is to obtain the spectral characteristics of a number of common combustion products under a variety of accurately known thermodynamics and optical conditions. Considerations leading to the design and fabrication of an experimental apparatus for this purpose are given in reference 1. This equipment consists of a graphite resistance furnace with an inert ceramic tube liner for the containment of high-temperature gases, and an optical system for detection of the infrared absorption. The test optical path is limited to the central zone of the furnace by the use of window holders at each end. Hot-pressed zinc selenide windows were originally employed because this material is transparent in the $1\text{-}20\ \mu$

wavelength region and inert to H_2O , CO_2 and CO up to a temperature of 540°F . In order to keep these windows below this limit it was necessary to water cool the holders in which they were mounted.

Data for the fundamental band of CO at temperatures of 300°K and 1800°K and at pressures from 0.25 to 3.0 atmospheres obtained with this system were reported in reference 1. Although agreement with existing results was within 15 per cent, the substantial temperature variation along the test optical path caused by the water-cooling of the windows made the comparison open to some question. Because of this the water-cooled window holders were replaced with alumina tubes and sapphire used for the windows in place of zinc selenide.

These modifications are described in this report. Measurements of the temperature along the test path showed the variation to be less than 100 K at 1200 K. Absorption measurements for CO using both the water-cooled and ceramic window holders are also presented and discussed.

INFRARED SPECTRAL ABSORPTION SYSTEM

The infrared spectral absorption system constructed for this investigation has been described in detail in reference 1. The hot-pressed zinc-selenide windows* located at the ends of the test optical path are inert to H_2O , CO and CO_2 in the temperature range up to 550°K . This temperature

* Eastman Kodak Irtran 4.

limitation made it necessary to water-cool the holders in which the windows were mounted. The large amount of cooling necessary to maintain these windows below 550 °K resulted in a large temperature variation through the optical test path. Typical temperature measurements in CO gas are presented in Fig. 13 of reference 1.

To eliminate this temperature variation, high temperature window holders were fabricated and mounted in the furnace as shown in Fig. 1. Each holder was made* from a high purity alumina tube (1" outside diameter, 1/8" wall and 6" long) with a sapphire window (approx. 1/8" thick) bonded in one end by using a high temperature glass frit. These sapphire windows (single crystals of pure Al_2O_3)** are transparent in the 0.2 - 7 μ wave-length region, are inert to H_2O , CO, CO_2 and can withstand temperatures up to 2000 °K.

The outer end of each alumina tube is bonded to a kovar tube by a copper braze. This kovar tube is threaded to an adjustable stainless steel cylinder as shown in Fig. 1, and is sealed to it with a crushable copper washer.

The glass frit seal mentioned above is amorphous in nature and weakens at high temperatures. To increase the operating range provisions were made

* By Eitel-McCullough Inc., 301 Ind. Way, San Carlos, California.

** Obtained from Linde Crystal Product Division, Union Carbide Corp., 499 Hamilton Avenue, Palo Alto, Calif.

to balance the pressures on both sides of the sapphire windows. This was accomplished by installing a second window (Na. Cl) at one end of the stainless steel cylinder as shown in Fig. 1. During operation, the pressures of inert gas in the regions between the Na. Cl windows and the sapphire windows are maintained equal to that of the test gas.

Measurements made with this system are thus limited to the highest service temperature of the glass frit (about 1500 °K) and to the transparent range of the sapphire windows.

EXPERIMENTAL RESULTS

Temperature Distributions

Temperature distributions along the optical test path with non-cooled alumina window holders and with water-cooled window holders are showed in Fig. 2. As can be noted the desired isothermal condition is almost achieved with the alumina window holders. The deviation from the maximum center temperature is reduced to within 80 °K when the center temperature is 1200 °K.

Absorption Measurements

Spectral absorptivities for the fundamental band of pure CO gas at a center temperature of 1200 °K, a path length of 10 cm, and pressures of 0.25 to 3 atm with alumina window holders and water-cooled window holders are shown in Figs. 3 and 4, respectively. In each case the temperature at the center (as measured with a thermocouple) was adjusted to 1200 °K. The procedure followed in the calculation of the spectral absorptivities was the same, as described in reference 1, (page 19). A comparison of the results

in Fig. 3 with those of Fig. 4 shows that the absorptivities with the water-cooled window holders are higher in the wave number region near the band origin but lower in the wave number region far from the band origin. This is due to the fact that the effective temperature along the test path length of CO gas with the water-cooled window holders is lower than that with the alumina window holders. An explanation for this is given in the next section.

DISCUSSION

Infrared radiation from gases results primarily from transitions in the vibration-rotation energy levels of the molecules which possess an electric moment (e.g., CO, CO₂, and H₂O). Separations of the rotational energy levels due to the interaction of vibration and rotation are so much smaller than those of the vibrational energy levels that the rotational lines in each vibrational band can only be distinguished by a high resolution spectrometer.

For radiative transfer studies such detailed observations are not practical and less sensitive spectrometers can be employed. However, interpretation of the measurements requires the application of an appropriate model to simulate the "fine structure" of the spectra, i.e., vibrational bands consisting of closely spaced rotational lines broadened primarily by molecular collisions. These models specify the band structure - intensity distribution, line-spacing distribution, and line-width distribution - and yield relations between absorptivity and band parameters, i.e., integrated band intensity, average line-spacing, and average line-width (see reference 2).

Correlation of Results

Attempts to correlate the data in Fig. 3 by employing either Lambert-Beer's law (essentially the weak-line approximation) or a statistical band model were not successful. This led to the consideration of the strong line approximation in the Elsasser band model which yields³

$$A = \operatorname{erf} \left[\left(\frac{1}{2} \beta^2 x \right)^{1/2} \right] \quad (1)$$

$$\text{where } \beta = \frac{2\pi \alpha^0}{d} P_e, \quad x = \frac{S}{2\pi \alpha^0} u_e,$$

$$\alpha = \alpha^0 P_e \quad u_e P_e = u = p\ell,$$

$$P_e = D p + \sum_i F_i p_i,$$

Here d is the line-spacing, P_e the effective total pressure, S the line intensity, u_e the effective amount of absorber, α the line-width, u the amount of absorber, p the partial pressure of absorbing gas, ℓ the geometric path length, p_i the partial pressure of the i^{th} foreign gas, D self-broadening constant, and F_i broadening constant due to the i^{th} foreign gas. Substitution of β and x into Eqn. (1) gives

$$A = \operatorname{erf} \left[\left(\frac{\pi S \alpha^0}{d^2} \right)^{1/2} (P_e u_e)^{1/2} \right] \quad (2)$$

In the present case, $p_i = 0$, and $D = 1.02$ (D for CO gas at 1200 °K is assumed to have the same value as that at room temperature). Thus P_e and u_e can be determined, and then $(\pi S \alpha^0/d^2)^{1/2}$ can be calculated from

the values of A in Fig. 3 by using Eq. (2). The results are presented in Fig. 5. Good correlation of the values for $(\pi S \alpha^0/d^2)^{1/2}$ with a mean line is obtained for the pressure range from 0.5 to 3 atm for the path length tested. This correlation however only serves to show the consistency of our data and the applicability of the strong line approximation in the Elsasser model for the range of conditions covered by the present experiment. Investigations over a wide range of conditions are necessary to establish the general validity of this model.

Comparison with Davies' Results

For purposes of comparison Davies' results⁴, at 2119 cm^{-1} , 2143 cm^{-1} and 2174 cm^{-1} were analyzed in the same way to obtain the values of $(\pi S \alpha^0/d^2)^{1/2}$. ($\ell = 7.62 \text{ cm}$; $F_{\text{argon}} = 0.78$; the concentration was taken as 0.40). The points obtained are plotted in Fig. 5 and can be seen to fall about 20% higher than the present results. This could be due to the possibility that the deviations from the strong line approximation may occur at the lower range values of the optical depth existing in Davies' experiments ($u \approx 3$ as compared to $u = 10$ in the present case).

Effect of the Non-Isothermal Path Length

Radiative absorption in a non-homogeneous gas has been extensively discussed by Plass.⁵ Absorption in a non-isothermal gas is to be treated as a special case of this general problem. When the strong line approximation in the Elsasser band model is applicable, the absorptivity for a non-homogeneous gas can be considered to be equal to the absorptivity

of an equivalent homogeneous gas if the following condition is satisfied

$$S_h \propto u_h = \int_0^u S \propto du' \quad (3)$$

Here the subscript h denotes a homogeneous gas.

To obtain an approximate expression for the integral in equation (3) we note that for pressure-broadened gases under constant pressure,

$$\alpha \propto T^{-1/2} \quad (4)$$

and for ideal gases,

$$\Delta u, \propto T^{-1} \Delta \ell \quad (5)$$

Also for a fundamental band whose integrated intensity is temperature independent,

$$S \propto \frac{1}{T} \exp [-BJ (J+1) hc/kT] \quad (6)$$

where J is the rotational quantum number and is an increasing function of $|\nu - \nu_0|$, B the rotational constant (equal to 1.922 cm^{-1} for CO), ν the frequency and ν_0 the frequency at band origin. Therefore we find

$$S \propto du \propto T^{-5/2} \exp (-\gamma/T) d\ell \quad (7)$$

where γ is an increasing function of $|\nu - \nu_0|$. Eq. (7) shows that $S \propto du/d\ell$ is a decreasing function of temperature for small γ and an increasing function of temperature for large γ . The temperature of the CO gas in the test path with water-cooled window holders is lower (except at the center) than that

with alumina window holders over the whole optical path. Therefore, in comparison with values of $\int_0^u S \propto d u'$ for the alumina window holders, the values of $\int_0^u S \propto d u'$ for the water-cooled window holders will be higher for ν near the band origin and lower for ν far from the band origin. Since the absorptivity is an increasing function of $S \propto u$, the absorptivity for gases with the water-cooled window holders is higher than that with alumina window holders near the band origin and vice versa. This is confirmed by comparison of the curves in Figs. 3 and 4.

Project Schedule

Immediate attention will be devoted to obtaining absorption data for CO over the maximum possible temperature, pressure, and path length ranges of the infrared system. The possibility of gas cooling of the alumina window holders to permit operation up to 2000 °K is being investigated.

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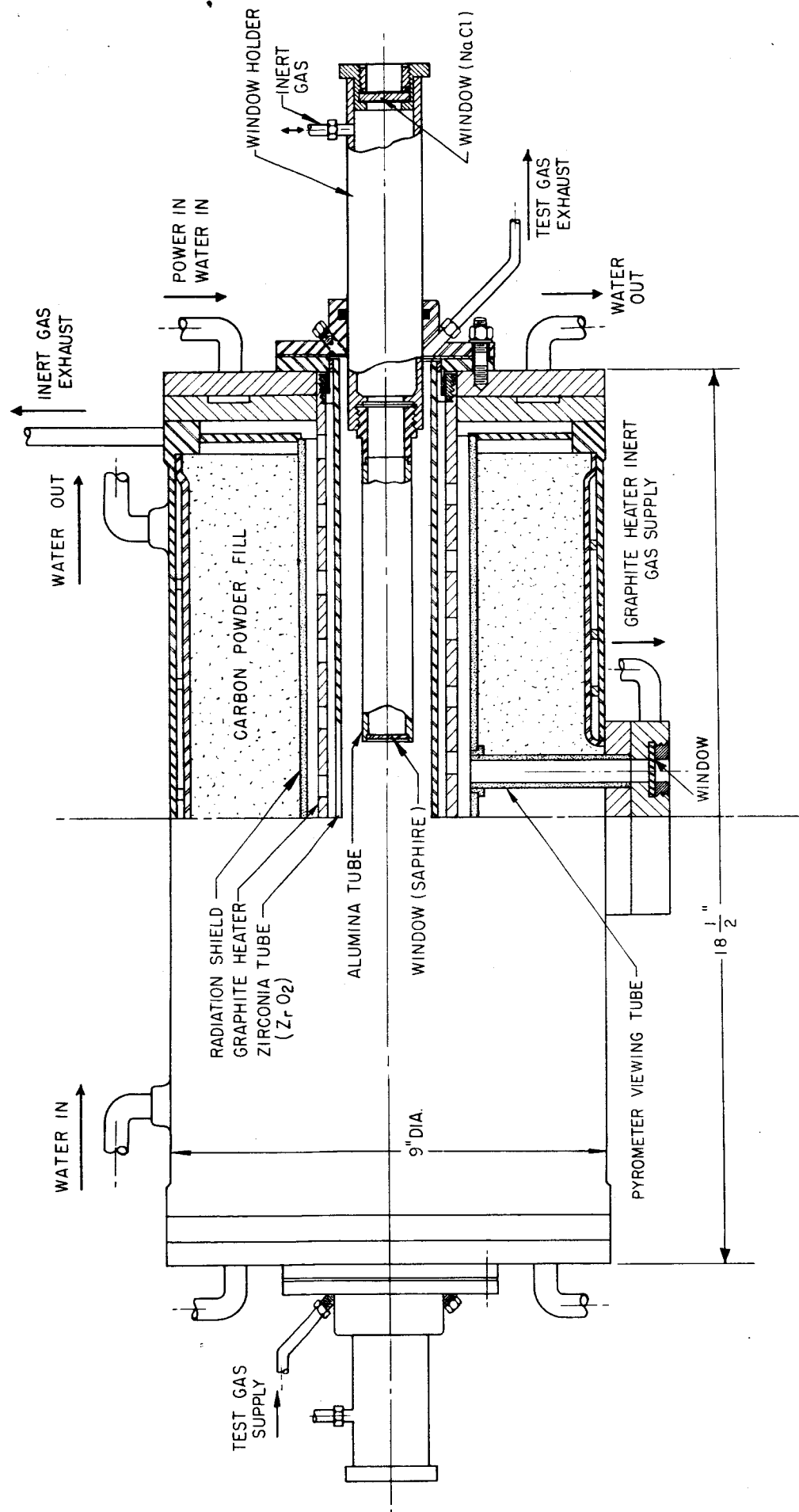


FIG. 1 HIGH TEMPERATURE FURNACE

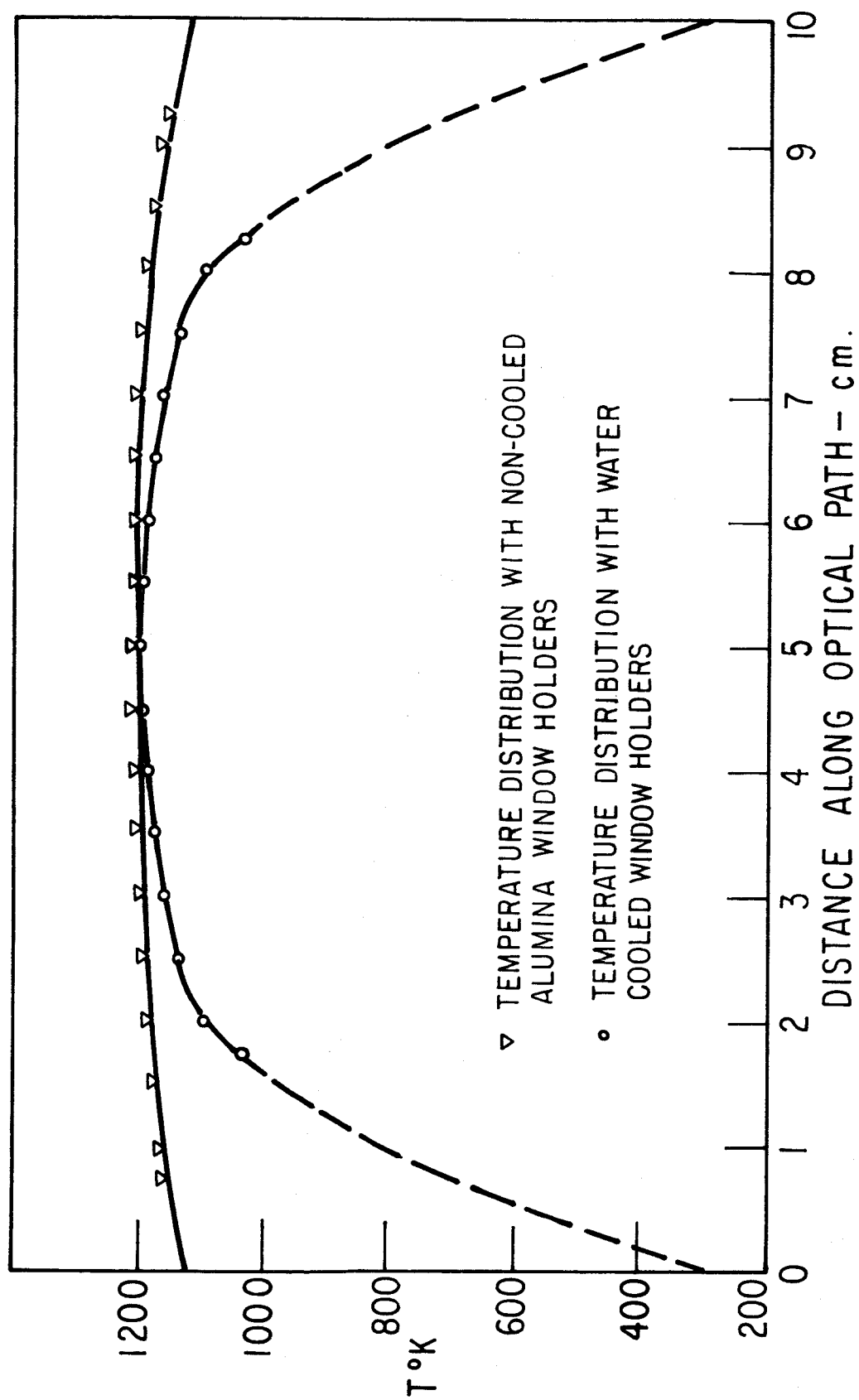


FIG.2 GAS TEMPERATURE DISTRIBUTIONS

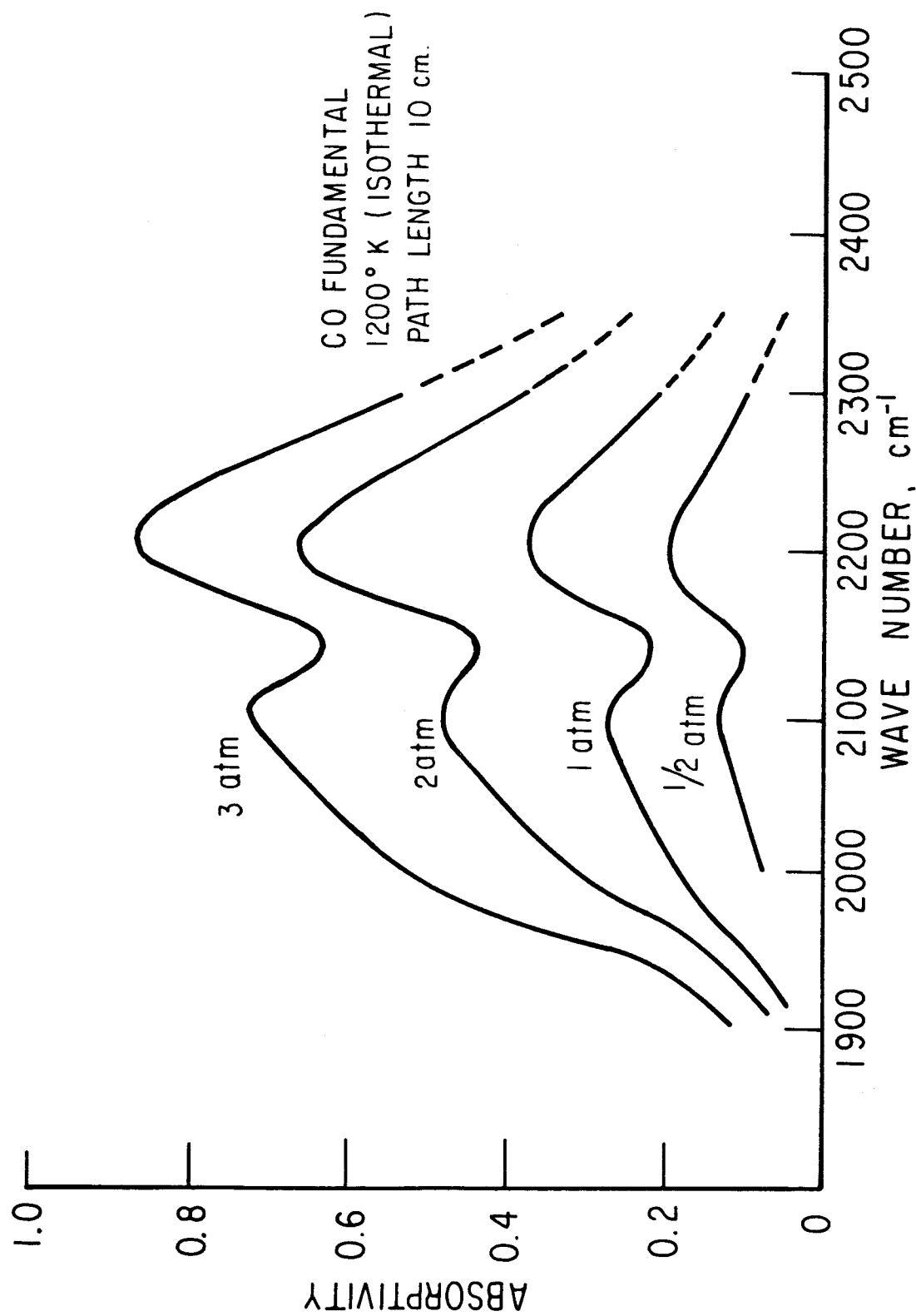


FIG.3 SPECTRAL ABSORPTIVITY OF FUNDAMENTAL BAND OF CO GAS AT TEMP. 1200° K AND AT PATH LENGTH 10 cm, USING CERAMIC WINDOW HOLDERS.

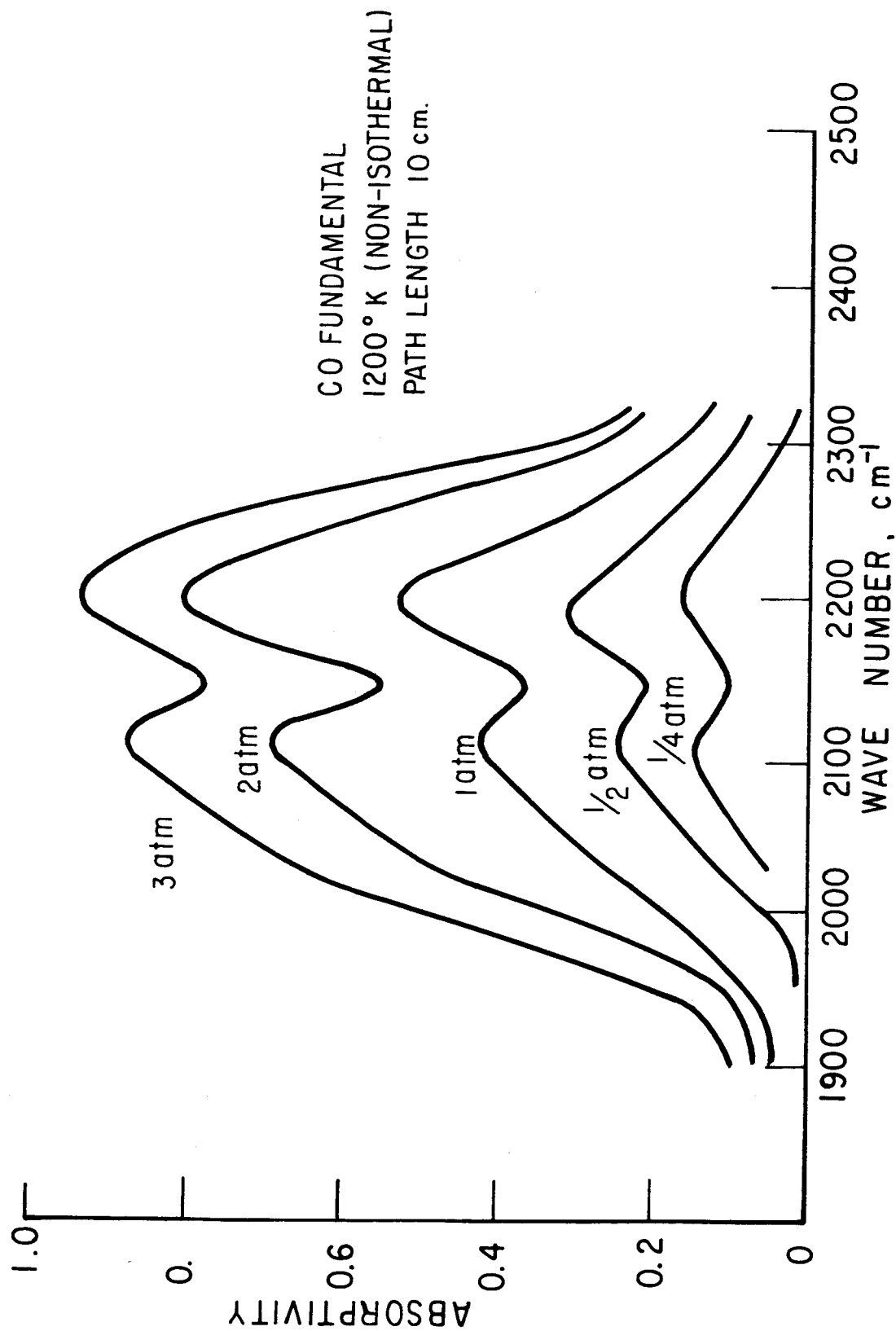


FIG.4 SPECTRAL ABSORPTIVITY OF FUNDAMENTAL BAND OF CO GAS AT CENTER TEMP.
1200°K AND PATH LENGTH 10 cm, USING WATER-COOLED WINDOW HOLDERS

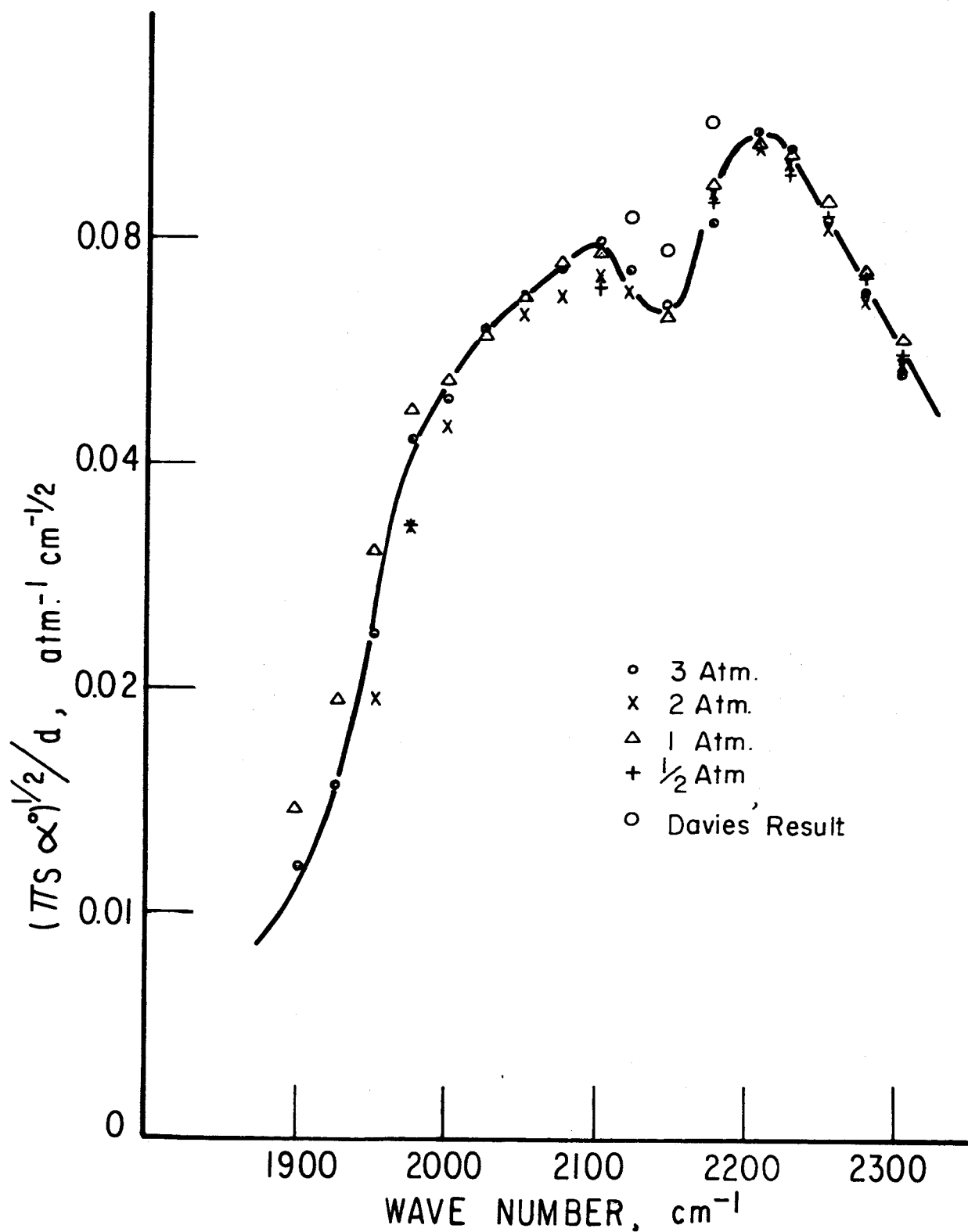


FIG. 5 $(\pi S \alpha^\circ)^{1/2} / d$ vs WAVE NUMBER FOR CO FUNDAMENTAL AT TEMPERATURE 1200 °K